Scientific Progress Report On DOE -NSSA
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First Principles Investigations of Americium, Plutonium and their Mixtures using Dynamical Mean Field Theory.

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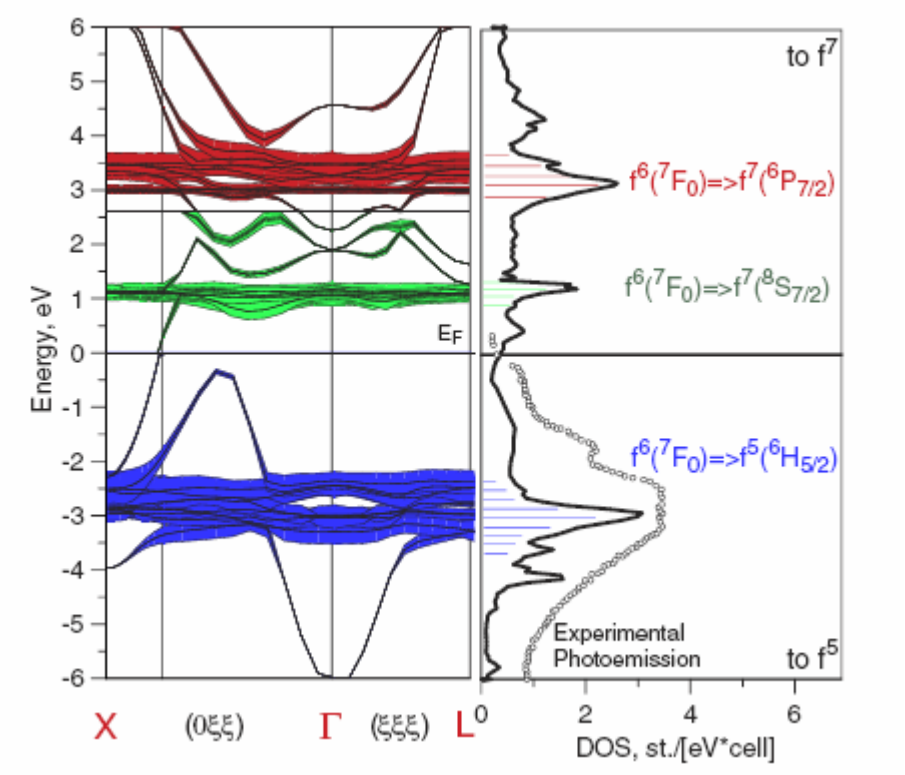
RESEARH ACCOMPLISHMENTS

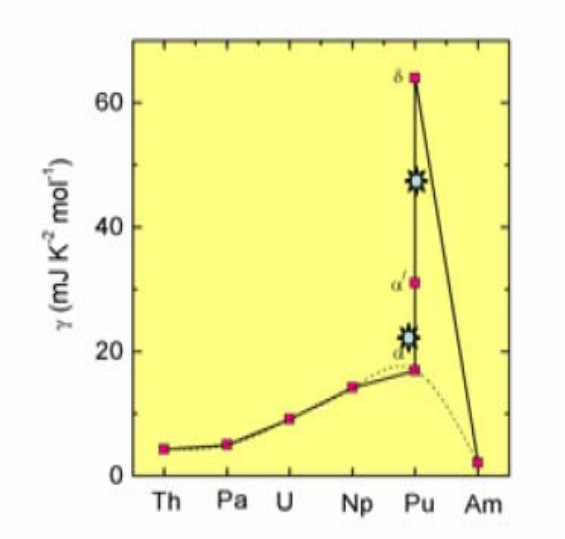
The ultimate goal outlined in our proposal is to investigate Am, Pu, and Am-Pu mixtures, to gain a basic understanding of their electronic structure, the basic principles controlling their physical properties, and to develop theoretical tools with predictive power for these late actinides. Using a relativistic electronic structure method based on dynamical mean field theory (DMFT) . All the objectives for the first year have been met. We detail the achievements below.

The first issue tackled was the first principles computation of the photoemission spectra of Americium metal. For this purpose we developed a new combination of DMFT with a Hubbard I solver. The results of the computation of the photomeission spectra are shown below. The method was validated by comparison with experimental data.



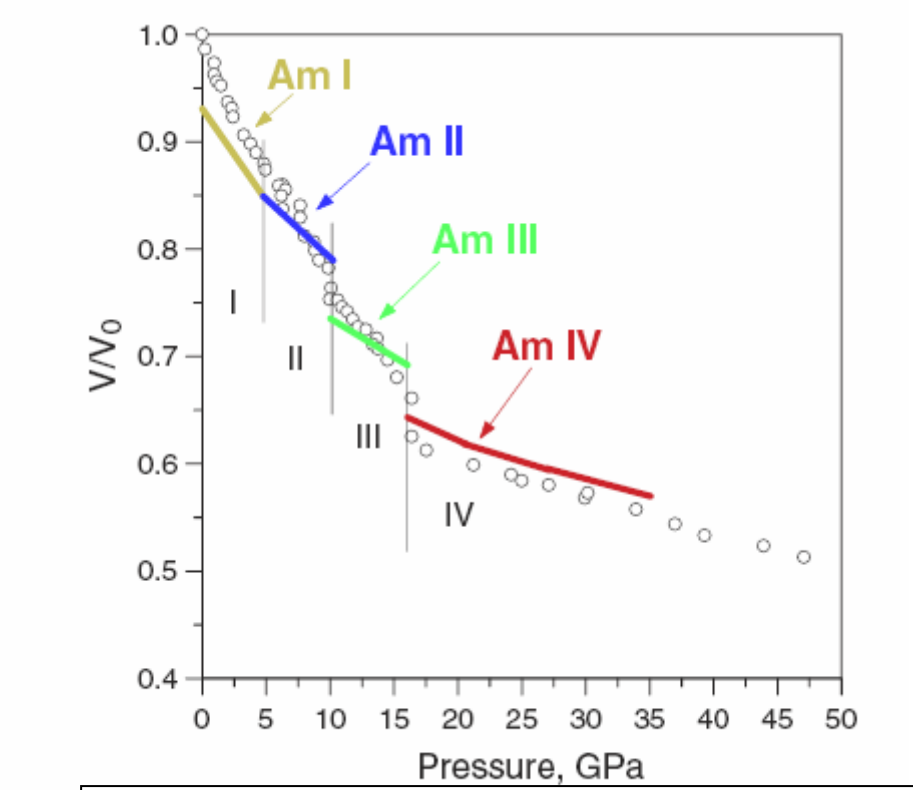
Angle resolved Photoemission and inverse photoemission spectra (left) and angle integrated photoemission spectra (right) compared with experimental data from ITU
The figure indicates the main electronic transitions

The second issue tackled was the evaluation of the linear term of the specific heat coefficient in alpha and delta plutonium. This is an important problem because they correlate with the approach to the Mott transition as indicated in the figure below from the work of Lashley et.al (PRB) .
As a first pass at this problem we used a simplified form of the structure of alpha plutonium, proposed by Bouchet et. al. and the LANL group of Bob Albers. The DMFT results are in excellent agreement with experimental data of alpha Pu, but not with delta Pu. Further studies are needed to see if the inclusion of multiplets in the delta structure improves the agreement with experiments and they will be undertaken in the forthcoming year.



Experimental data for the specific heat of delta and alpha plutonium DMFT FLEX (flex) computations with a simplified structure are in good agreement for alpha Pu . The same method slightly underestimates the specific heat of delta Pu . as described in ref 2. The experimental points are from Lashley et. al. the stars are the theoretical points

Finally we undertook the evaluation of the equation of state at zero temperature and very high pressures of Americium metal. For this purpose we developed a new algorithm to solve the LDA+DMFT equations. In this approach, we introduced a generalization of Kohn Sham eigenvalues and eigenstates which are appropriate for the description of the Hubbard bands. It turns out that the introduction of these quantities speeds up the total energy computations by a factor of 10, allowing us to evaluate for the first time total energies pressures and equations of state of complex structures with many atoms per unit cell. The comparison of our results with experiments is shown in figure below.



Comparison of the theoretical DMFT Hubbard I equation of state with experimental data of ITU The agreement is quite satisfactory. Our theory is published in ref 1

Deliverables: we have develop a code to evaluate the total energy using DMFT Hubbard I at fixed value of the impurity level.

Publications During the First Year of Funding:

Ref 1 Many-Body Electronic Structure of Americium metal: Sergej Y. Savrasov, Kristjan Haule, Gabriel Kotliar, PRL 96, 036404 (2006)

Ref 2 L. Pourovski G. Kotliar M. Katsnelson and S. LichtesteinAn dynamical-mean-field-theory investigation of specific heat and electronic structure of alpha and delta plutonium arXiv:cond-mat/0702342 :

COST STATUS:

There are no underspent funds.

Collaborations:

There is a close collaboration between the PI's G. Kotliar and S. Savrasov and postdocs Alex Gordienko, and K. Haule a junior faculty at Rutgers, as well as L. Pourovski and postdoc and two senior researchers, A. Lichtenstein and M. Katsnelson in Europe.